

1 The role of groundwaters in the global carbon cycle: Insights 2 from the contiguous United States

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7 Supplement Text

8 DATA COMPILATION

9 Initially, we collected HCO_3^- from 159,319 wells and 38,103 river gauges, and discharge
10 rates from 7,199 river gauges, across the continental United States from USGS archives. After
11 data collection, we only selected samples that were analyzed following the “sample-routine”
12 procedure. We further filtered out the samples that do not have reported values. Several types of
13 streams are denoted by USGS, including ditch, canal, perennial stream, tidal stream, channelized
14 stream, intermittent stream, ephemeral stream, pond-stormwater, and normal stream. To mitigate
15 the human intervention on the river chemistry, we only selected the normal stream and the
16 perennial stream as our research target. For both groundwaters and rivers, we further selected
17 samples that have unique activity identifiers to avoid sampling ambiguity. As the bicarbonate
18 concentrations for both groundwaters and rivers are extremely right skewed, we used 99%
19 quantile as a cutoff to get rid of the extremely large values. We further selected the wells and
20 river gauges that have at least one data point in each season, from which the annual bicarbonate
21 concentration for each well and river gauge, and the annual discharge rates for each gauge, were
22 calculated. We also joined the dataset of bicarbonate concentration and the dataset of discharge

rate at each gauge together. After tidying the data, we finally have 4258 data points for river and 3131 data points for groundwater.

[HCO₃⁻] IN GROUNDWATERS ALONG THE COASTLINE OF CONTIGUOUS UNITED STATES

As most groundwaters will eventually discharge into ocean along the coastline, we further investigated the spatial distribution of HCO₃⁻ in groundwaters by creating four buffer zones close to the coastline — a 5 km buffer, a 10 km buffer, a 25 km buffer and a 50 km buffer (Fig. DR3A). The statistical analysis shows that in all of these buffer zones, there is elevated groundwater [HCO₃⁻] in comparison with rivers, with Monte Carlo resampled means ranging from 186 to 215 mg/L for groundwater compared to 60 to 99 mg/L for river. The coastal groundwater/river [HCO₃⁻] ratio (2.2 to 3.2; Fig. DR3B) is similar across coastal buffer zones to the entire contiguous United States groundwater vs river [HCO₃⁻] ratio (2.4 in the main text).

MEAN [HCO₃⁻] IN GROUNDWATERS OF ADDITIONAL LOCATIONS BESIDES THOSE MONITORED BY USGS

To achieve a more robust estimate of global mean groundwater [HCO₃⁻], we collected additional groundwater data from other locations in the US and other regions (Table DR1). These data points are from North Inlet of South Carolina (Cai et al., 2003), Okatee Estuary of South Carolina (Moore et al., 2006), Martinique and Guadeloupe volcanic island, Reunion volcanic island (Rad et al., 2007), northern South China Sea coastline (Liu et al., 2012), the Baltic Sea coastline (Szymczycha et al., 2014), Godavari estuary from India (Rengarajan and Sarma, 2015) and Taiwan coastal areas (Chen et al., 2018; Wang et al., 2018). Some studies only

list the concentration of dissolved inorganic carbon (DIC) in groundwater and we have to transform them to $[HCO_3^-]$. For groundwater samples, the dominant carbon species are usually $[HCO_3^-]$, especially at $pH < 8.5$ (Schopka and Derry, 2012). This is further supported by the studies by Chen et al. (2018) and Wang et al. (2018), which analyzed the same batch of groundwater samples and showed that $[HCO_3^-]$ amounts to ~94% of DIC (Table DR1). To make a conservative estimation, we multiplied the DIC in the relevant study with 90% to obtain a first-order $[HCO_3^-]$. After transformation, the $[HCO_3^-]$ in the new groundwater samples ranges from 3.0 mmol/L to 7.1 mmol/L, with an exception of one sample with 14.4 mmol/L from Okatee Estuary of South Carolina. These new estimates are comparable to the mean of $[HCO_3^-]$ in contiguous US (4.4 mmol/L). To make a more conservative estimation, we selected the majority range (from 3.0 mmol/L to 7.1 mmol/L) to sample from in order to obtain the global groundwater $[HCO_3^-]$.

“ONE-STEP APPROACH” AND “TWO-STEP APPROACH” FOR ESTIMATING FRESH SUBMARINE GROUNDWATER DISCHARGE

In this study, we adopted two distinct approaches to estimate the fresh submarine groundwater discharge (SGD). The first one is numerical modeling, which we call “One-step approach” as it is a single calculation to estimate fresh SGD. The second one is empirical measurement, which we call “Two-step approach” as it needs to estimate both the mixed SGD and the proportion of fresh SGD in mixed SGD in order to obtain the fresh SGD.

The “One-step approach” can be further divided into two numerical modeling methods: the water balance model and the groundwater flow model. The water balance model calculates groundwater discharge by considering the balance between the recharge rate, export rate to

streams, evapotranspiration losses and other processes for a specific recharge area (Sawyer et al., 2016; Zhou et al., 2018, 2019). The groundwater flow model utilizes the calibrated hydraulic properties, hydrologic connections, stratigraphic data, aquifer properties, and recharge rates, etc (Befus et al., 2017; Zhou et al., 2018; Luijendijk et al., 2019). It is more complex than the water budget model and can illustrate flow paths and fresh groundwater contributions from heterogeneous groundwater systems (Zhou et al., 2018).

The old estimates based on the “One-step approach” yielded a global fresh SGD as 6% (Zektser et al., 2007) or 10% (Garrels and Mackenzie, 1971) of the global river discharge. For the study of Zektser (2007), Moore (2010) argued that it might underestimate the fresh SGD as it only includes fresh SGD from the shallow zones of the recharge zone and might miss fluxes from deeper zones. Using a water balance model, Sawyer et al. (2016) argued that the fresh SGD is equal to <2% of the river discharge for the US coastline. Based on a groundwater flow model, Befus et al. (2017) proposed that the fresh groundwater discharge to the eastern US and Gulf of Mexico coastline amounts to 13% of the river discharge. Zhou et al. (2018) conducted model comparison (water balance model and groundwater flow model) and inferred that the fresh SGD along the eastern US and Gulf of Mexico coastline is ~6% of the river discharge. Later, using a water balance approach, Zhou et al. (2019) determined a near-global fresh SGD to be ~1.3% of the global river discharge. The main factor leading to these different estimates—and the largest uncertainty in this approach overall—is the selection of a recharge zone for the groundwater flow regime. Larger recharge zones (Befus et al., 2017; Zhou et al., 2018) (using HUC8 or HydroSHEDS) generally lead to higher estimate while small recharge zones will yield lower estimate (Sawyer et al., 2016) (using NHDPlus). To sum up, those recent studies imply the fresh SGD is ~1.3%–13% of the river discharge on the continental or global scale, similar to the old

estimates of 6%–10%. It is also noted that one study, based on groundwater flow model, argues for a much lower fresh SGD, which is suggested to be 0.001%–0.6% of river discharge (Luijendijk et al., 2019). However, some of the results from this study are in conflict with local fresh SGD estimates. This approach also has to assume a recharge zone for the groundwater flow. Nonetheless, we still treated this study as one end member scenario, which highlights the large uncertainty in the current estimate of fresh SGD. To summarize, for the global estimates using the “One-step approach”, we chose a range of fresh SGD to be 1.3%–13% of river discharge in the normal scenario (“Groundwater_one_step” in Fig. DR5), and 0.001 % to 0.6% of river discharge in the second scenario (“Groundwater_one_step_small” in Fig. DR5). The fresh SGD estimates using the “One-step approach” could be found in Table DR2.

The “Two-step approach” is a combination of the tracer approach and other measurements or modeling. The tracer approach is used to determine the total mixed SGD (Kwon et al., 2014; Cho and Kim, 2016). The principle of the tracer approach is straightforward. As radium (^{226}Ra and ^{228}Ra) is highly enriched in salty coastal groundwater relative to the ocean and river, small input of groundwater into the ocean can lead to a strong signal. Therefore, with the knowledge of radium isotope values in different components (e.g., river, ocean, suspended particle, sediment advection flux, groundwater) and the fluxes of different components (except for groundwater), one could obtain the groundwater contribution to the radium in coastal water. The short-lived radium isotopes (^{223}Ra and ^{224}Ra) can be used to evaluate the residence time or mixing rate of coastal waters (Moore et al., 2006) and further could be used to estimate radium fluxes from the ocean side. ^{222}Rn (The daughter of ^{226}Ra) is highly enriched in both fresh and salty groundwater, and could also be useful in estimating the groundwater fluxes. Typically, based on these tracers, we will obtain the total flux of groundwater (fresh component and

modified seawater component). By integrating global observations of ^{228}Ra with an inverse model, Kwon et al. (2014) argued that the mixed SGD in the coastlines between 60°S and 70°N is $9\text{--}15 \times 10^{13} \text{ m}^3/\text{yr}$, which is ~ 3 times greater than the river discharge. In contrast, Cho and Kim (2016) stated that instead of 3 to 4 times, the global mixed SGD is approximately 90% to 175% of the river discharge, yielding a flux of $3.2\text{--}5.6 \times 10^{13} \text{ m}^3/\text{yr}$.

To estimate the proportion of fresh SGD in total SGD, we need to resort to specific seepage measurement, conductivity measurement, numerical modeling, stable isotope measurements, and a multi-component tracer approach (Garrison et al., 2003; Kim et al., 2003; Taniguchi and Iwakawa, 2004; Povinec et al., 2006; Taniguchi et al., 2006, 2008; Mulligan and Charette, 2006; Hays and Ullman, 2007; Kroeger et al., 2007; Beck et al., 2007; Niencheski et al., 2007; Sieyes et al., 2008; Leote et al., 2008; Santos et al., 2009; Lee et al., 2012; Russoniello et al., 2013; Null et al., 2014; Rodellas et al., 2015; Szymczycha and Pempkowiak, 2016; Sadat-Noori et al., 2016; Petermann et al., 2018; Chaillou et al., 2018; Chen et al., 2018; Adyasari et al., 2019). Note that the proportion of fresh SGD can also be estimated through numerical modeling (Li et al., 1999). The studies (Fig. DR4) estimating the proportion of fresh SGD in the mixed SGD range from 1% to 97%. Here we chose the majority results ranging from 1% to 50%. Multiplying the conservative global mixed SGD estimated by Cho and Kim (2016) with the proportion of fresh SGD in the mixed SGD, we arrived at a fresh SGD to be 1% to 77% of the total river discharge. The literature review on the total SGD and proportion of fresh SGD could be found in Table DR2.

With the result of global fresh SGD either from the “One-step approach” or from the “Two-step approach”, we could simply multiply this value with the global estimate of groundwater $[\text{HCO}_3^-]$ to arrive at the global fresh SGD HCO_3^- flux. We note that the small end

member in the “One-step approach” (“Groundwater_one_step_small”; Fig. DR5) predicted a much smaller value for both the global fresh SGD HCO_3^- flux (2.68×10^{11} (25th percentile)– 8.15×10^{11} (75th percentile), which is 1%–2% of the river flux) and the flux derived from silicate weathering (1.60×10^{11} – 4.76×10^{11} , which is 1%–3% of the river silicate flux). It is hard to judge whether this end member reflects the truth given the larger estimates predicted by the recent studies (normal scenarios; “Groundwater_one_step”; Fig. DR5) and the potential underestimation by this study. However, it does highlight that there are relatively large uncertainties in the extent of groundwater carbon sink, which provides a strong impetus to further constraining the extent of SGD on a global scale.

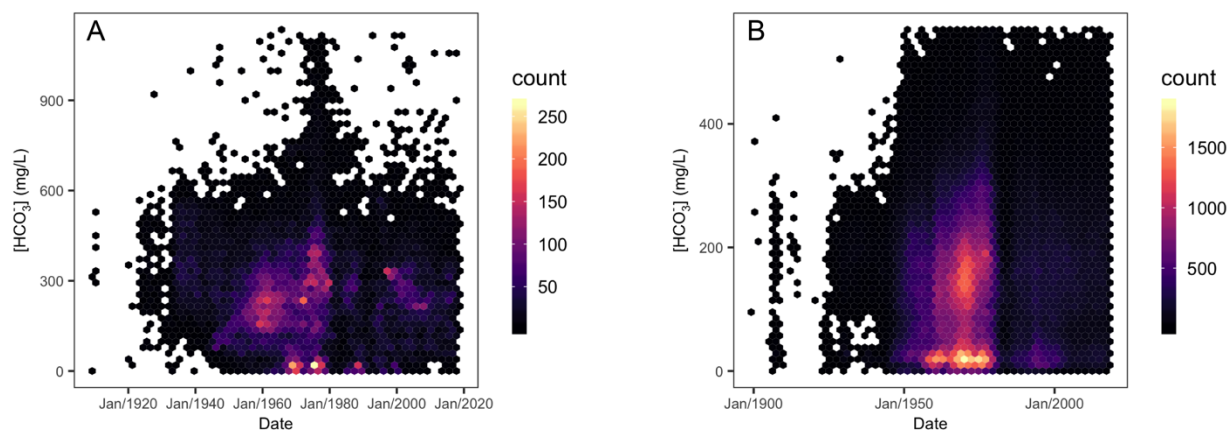


Fig. DR1 Density of HCO_3^- concentration ($[HCO_3^-]$) as a function of the time across the contiguous United States. (A) Groundwater samples (B) River samples.

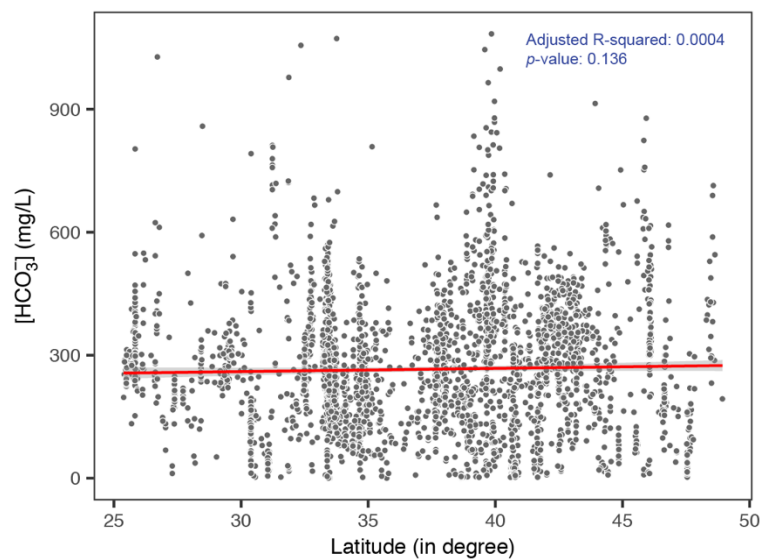


Fig. DR2 HCO_3^- concentration ($[HCO_3^-]$) in the groundwater as a function of the latitude of the sampling location across the contiguous United States.

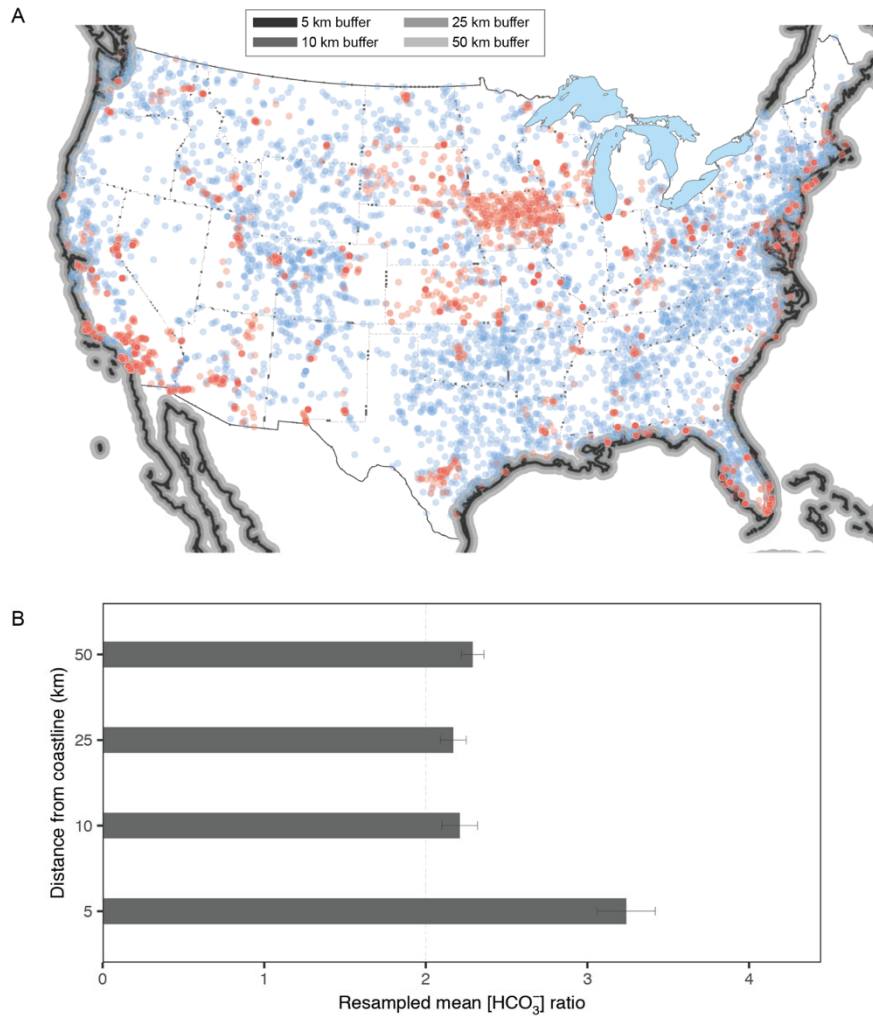
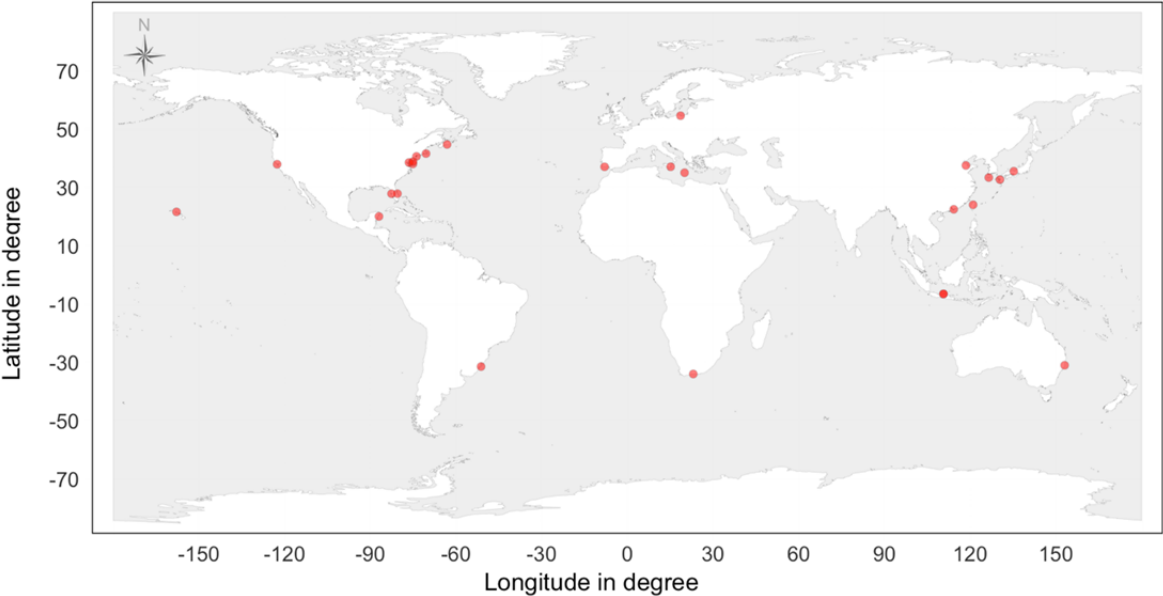


Fig. DR3 Buffer zone analysis for the groundwater and river samples. (A) Locations of river gauges and groundwater wells across the contiguous United States. Blue dots represent river samples and red dots represent groundwater samples. From dark grey to light grey, the width of the buffer zone increases from 5 km, through 10 km and 25 km, to 50 km. (B) Resampled mean $[HCO_3^-]$ ratio of groundwater vs river located in the buffered zones of the US coastline. From bottom to top, the width of buffer zone increases from 5 km, through 10 km and 25 km, to 50 km. Error bars represent 1 SD for the resampled mean ratio.

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171 Fig. DR4 Locations that have the data for the proportion of fresh SGD in the total mixed SGD.

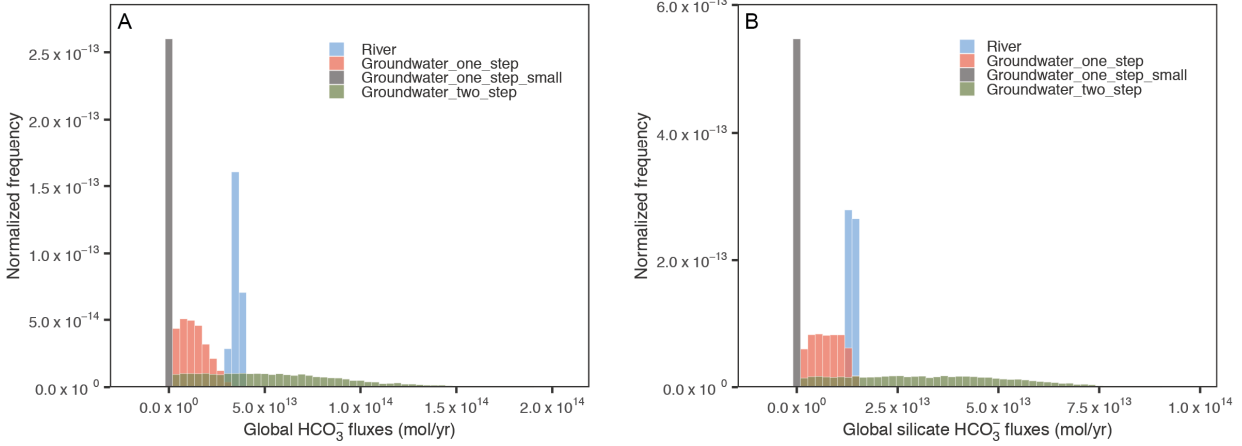
172 The locations can be found in Table DR2.

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178 Fig. DR5 Global estimates of HCO_3^- fluxes from groundwaters and rivers. (A) Normalized
 179 frequency of global HCO_3^- fluxes from groundwaters and rivers. (B) Normalized frequency of
 180 global HCO_3^- fluxes from groundwaters and rivers in the silicate watersheds. The groundwater
 181 estimates are based on the “One-step approach” and the “Two-step approach”. Blue color
 182 represents river, red color represents the groundwater estimate using the “One-step approach”,
 183 grey color represents the groundwater estimate using the small scenario of “One-step approach”,
 184 and green color represents the groundwater estimate using the “Two-step approach”.

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186 Table DR1: Data compilation for groundwater [HCO_3^-] around the world.

Parameter	Range (mmol/L)	Mean (mmol/L)	Mean HCO_3^- (mmol/L)	Location	Data source
HCO_3^-	0–17.7	4.4	4.4	Contiguous United States	This study
DIC	2–14.5	5.0	4.5	North Inlet, South Carolina	Cai et al. (2003)
DIC	16	16.0	14.4	Okatee Estuary, South Carolina	Moore et al. (2006)
HCO_3^-	0.095–13	3.5	3.5	Martinique and Guadeloupe volcanic island	Rad et al. (2007)
HCO_3^-	2.008–4.016	3.0	3.0	Reunion volcanic island	Rad et al. (2007)
DIC	4.125–6.625	5.4	4.8	northern South China Sea coastline	Liu et al. (2012)
DIC	0.73–9.36	5.1	4.5	the Baltic Sea coastline	Szymczycha et al. (2014)
DIC	2.2–19.6	7.9	7.1	Godavari estuary, India	Rengarajan and Sarma (2015)
HCO_3^-	0.54–8.25	3.0	3.0	Taiwan coastal areas	Chen et al. (2018)
DIC	0.36–8.675	3.2	3.0	Taiwan coastal areas	Wang et al. (2018)

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189 Table DR2: Data compilation for the “One-step approach” and “Two-step approach”

One-step approach of estimating fresh SGD				
Method	Value	Unit	Location	Data source
Water balance model	10%	percentage of river discharge	global	Garrels and Mackenzie (1971)
hydrogeological model	6% – 7%	percentage of river discharge	global	Zekts et al. (2007)
Water balance model	1.3%	percentage of river discharge	global	Zhou et al. (2019)
Water balance model	<2%	percentage of river discharge	U.S. coastline	Sawyer et al. (2016)
Groundwater flow model	13%	percentage of river discharge	Eastern U.S. and Gulf of Mexico	Befus et al. (2017)
Water balance model and groundwater flow model	5% – 6%	percentage of river discharge	Eastern U.S. and Gulf of Mexico	Zhou et al. (2018)
Spatially resolved density-dependent model	0.001% – 0.6%	percentage of river discharge	global	Luijendijk et al. (2019)
Two-step approach of estimating fresh SGD				
Step 1: Mixed SGD				
Method	Value	Unit	Location	Data source
Tracer	9 – 15	$10^{13} \text{ m}^3 \text{ yr}^{-1}$	global	Kwon et al. (2014)
Salinity-based tracer approach	3.2 – 5.6	$10^{13} \text{ m}^3 \text{ yr}^{-1}$	global	Cho and Kim (2016)
Step 2: Percentage of fresh SGD in total mixed SGD				
Method	Value	Unit	Location	Data source
Numerical simulation	4	%	global	Li et al. (1999)
Tracer	10	%	The Chesapeake Bay, U.S.	Hussain et al. (1999)
Integration of model and seepage	2 – 22	%	Jeju Island the South Sea of Korea	Kim et al. (2003)
Integration of seepage, tracer and salinity	16	%	Kahana Bay in Hawaii	Garrison et al. (2003)
Integration of model and seepage	1 – 29	%	Osaka bay, Japan	Taniguchi and Iwakawa (2004)
Integration of tracer and model	80	%	Waquoit Bay, Massachusetts	Mulligan and Charette (2006)
Stable isotope	40 – 50	%	south-eastern Sicily, Italy	Povinec et al. (2006)
Integration of conductivity and seepage	6.1 – 37.7	%	Kyushu island, Japan	Taniguchi et al. (2006)
Integration of seepage and salinity	31.5	%	Cape Henlopen, Delaware	Hays and Ullman (2007)
Model and tracer	36	%	Southern coast of Brazil	Niencheski et al. (2007)
Integration of tracer and model	3.3	%	Jamaica Bay, New York	Beck et al. (2007)
Integration of tracer and model	20 – 50	%	Tampa Bay, Florida	Kroeger et al. (2007)
Seepage	10 – 30	%	the Ria Formosa, Portugal	Leote et al. (2008)
Seepage and tracer	2 – 22	%	Bohai Sea, China	Taniguchi et al. (2008)
Analytical models and water budget analysis	1.3 – 16	%	Stinson Beach, California	de Sieyes et al. (2008)
Integration of tracer and salinity	3 – 12	%	Gulf of Mexico	Santos et al. (2009)
Integration of tracer and model	3	%	Tolo Harbour, Hong Kong	Lee et al. (2012)
Seepage	1.2 – 46	%	Indian River Bay, Delaware	Russoniello et al. (2013)
Model and tracer	2	%	Coast of the Yucatan Peninsula	Null et al. (2014)
Integration of tracer and model	1 – 25	%	Mediterranean Sea	Rodellas et al. (2015)
Geochemical methods	18 – 45	%	Bay of Puck along the Polish coast	Symczycha and Pempkowiak (2016)
Seepage and tracer	59	%	Subtropical estuary of Hat Head, Australia	Sadat-Noori et al. (2016)
Tracer and stable isotopes	30 – 97	%	Martinique Beach of Quebec	Chaillou et al. (2018)
Tracer	23	%	Knysna Estuary, South Africa	Petermann et al. (2018)
Integration of seepage and salinity	36	%	Taiwan coastline	Chen et al. (2018)
Tracer	40	%	Awur beach of Indonesia	Adyasari et al. (2019)
Tracer	3	%	Bandengan beach of Indonesia	Adyasari et al. (2019)

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